Response to UDEQ K_d Interrogatories

Prepared for:

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Date April 22, 1997 Report 3101B.970422



Innovative Environmental Solutions

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Fax Cover

To:

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From:

Susan Wyman

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Envirocare of Utah, Inc.

Date:

February 15, 1999

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George,

Attached is a fax summarizing our evaluation of the Kd for PCBs. The available data indicate that PCBs will be acceptable for disposal in the LARW cell, without question.

We believe that the attached information should adequately establish the immobility of PCBs in soil and groundwater. We do not expect that UDEQ DSHW will require additional work (testing or other research) to demonstrate these facts.

While our geochemical laboratory is more than capable of conducting batch Kd tests, we do not believe such tests are necessary in this case, and we do not recommend them.

Thank you for the opportunity to be of service. If you need additional information, please call me or Mark Williamson at any time.

- Susan Wyman

Adrian**Brown**

Technical Memorandum

To:

George Hellstrom, Envirocare of Utah, Inc.

From:

Mark Williamson, Susan Wyman

Date:

February 15, 1999

Subject:

PCB Waste In LARW Cell

Project No.: 3101G

The transport characteristics of polychlorinated biphenyls (PCB's) were evaluated to determine whether these constituents would be acceptable for disposal at the Envirocare facility. Our evaluation indicates that PCBs do not represent a threat to groundwater and should be acceptable for placement in the LARW cell, because the migration potential of PCBs is low and the design of the cell (based on previous modeling) is protective. Even without the protectiveness of the LARW cell, the chemical properties of PCBs are such that they are strongly adsorbed by solids phases rather than being partitioned to an aqueous (water) component.

The partition coefficient, K_d , for a chemical constituent numerically describes the extent to which the chemical will adsorbed to a solid, rather than the aqueous phase in contact with that solid. For organic compounds, like PCBs, the value of K_d may be calculated using either the organic carbon partition coefficient, K_{oe} , or the octanol-water partition coefficient, K_{oe} (U.S. E.P.A., 1996)

When using K_{∞} ,

$$K_{d} = K_{\infty} \cdot f_{\infty} \tag{1}$$

where f_{oc} is the organic carbon fraction of the solid phase (i.e. soil or sediment). In cases where the value of f_{oc} is low (e.g. less than 1%), the value of K_d can be estimated from K_{ow} to estimate simple adsorption onto non-carbon surfaces

$$\log K_{d} = 1.01 \log K_{ow} - 0.36 \tag{2}$$

It is also not uncommon to use K_{ow} in place of K_{oc} in Eqn. 1, as these values tend to be very similar in magnitude.

No data are readily available for the organic content of the sediments underlying the LARW cell, but it is expected to be very low. Assuming a value for f_{∞} of 0.01 (1%) and a typical K_{ow} value of 10⁶ (see tabulations in Dulfer and Govers, 1995;M°Groddy, et al., 1996; Mackay, et al., 1992; ORNL, 1987), we can calculate a K_d value

$$K_d = K_{oc} f_{oc}$$
 (3)
= $10^6 \cdot 0.01 = 10^4$

Similarly, using Eqn. 2

$$Log K_d = 1.01log K_{ow} - 0.36$$
 (4)

Adrian**Brown**

Technical Memorandum

$$= 1.01(5) - 0.36 = 4.69$$
$$= 10^{4.69}$$

Both of these estimates are very large(on the order of 10,000) and indicate that PCBs do not have a tendency to migrate in the environment. Thus, in conjunction with the engineering design of the LARW to limit infiltration of precipitation, the placement of PCB's in the LARW cell would appear to suitable and protective.

References

- Dulfer, W.J. and Govers, H.A.J. (1995) Membrane-water partitioning of polychlorinated biphenyls in small unilamellar vesicles of four saturated phosphatiddycholines. *Environ. Sci. Tech.* 29: 2548-2554.
- Mackay, D., Shiu, W.Y., Ma, K.C. (1992) Illustrate Handbook of Physical-Chemical Properties and Environmental Fate for Organic Chemicals. Lewis Publishers, Inc. Chelsea, MI. Volume I.
- McGroddy, S.E., Farrington, J.W. and Gschwend, P.M. (1996) Comparison of the *in Situ* and desorption sediment-water partitioning of polyaromatic hydrocarbons polychlorinated biphenyls. *Environ. Sci. Tech.*. 30: 172-177.
- ORNL, Oak Ridge National Laboratory (1987) Toxicological Profile for Selected PCB's (Aroclor-1260, -1248, -1242, -1232, -1221, and -1016. Prepared by Syracuse Research Corporation, 68-03-3228.
- U.S. E.P.A., United States Environmental Protection Agency (1996) Soil Screening Guidance: Technical Background Document. EPA/540/R-95/128. Washington, D.C.

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1. ORIGIN OF THE UNIT 3 SAND SAMPLES

Please provide a discussion of the origin of the Unit 3 Sand samples, including the site location and depth from which the samples were collected and an explanation of why they are representative or appropriate for use in site-specific measurements. Details should include, but are not limited to, soil particle gradation or grain size distribution; porosity, bulk density, and mineral content (both reactive and non-reactive). In addition, discuss the soil grain sizes used in the tests and justify why they lead to representative or conservative K_d results when compared to field conditions.

Unit 3 samples collected for testing came from surface excavations where overlying material had been removed. The specific locations are noted in the site map contained in the Bingham Environmental report of August 1995. Samples were collected as stated in the workplan, from at least one foot below contact. Grain size distributions are the only characterizing tests performed and indicate that the material collected was Unit 3 sand, similar to that found at or near the water table. Attached Figures 1, 2 and 3 show the comparison between material collected for this program and the Unit 3 material collected during installation of GW-19A. In Figure 3, one extraneous profile is present for a unit other than Unit 3 sand.

Grainsize distributions for Unit 3 solids used in K_d testing have a low proportion of clays relative to site-wide distribution for Unit 3. The relationship is illustrated in Figure 1 below. As clay minerals commonly have a significant sorption capacity, the use of low clay content materials during testing would have the expected effect of producing a lower K_d than would be expected for typical Unit 3 sand.

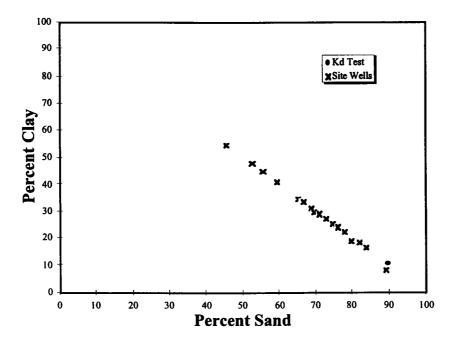


Figure 1. Clay fraction of Unit 3 sand.

No chemical or mineralogical analyses of the material used in the study, or for Unit 3 sands elsewhere on site, have been performed. However, geologic logs of materials collected during installation of monitoring wells does not indicate any dramatic color changes, such as iron staining, that would suggest a significant change in the chemical character of cements in the sands as a function of distance, laterally or with depth. The similarity of the grain size distribution charts suggest a consistent amount of fines (clays) in the sand matrix. Given that the geologic depositional environment would not be expected to produce dramatic gradients in clay mineral abundance, coupled with the significant period of time over which these materials have lain in the saturated brine groundwater to reach an equilibrium condition, suggests that dramatic changes in the clay mineral (or other minor mineral) content over the limited area defined by the LARW would not be expected. Thus, the solid material collected for K_d determinations would expected to be quite consistent with Unit 3 sands at depth in the system.

2. SELECTION OF GROUNDWATER SOURCES USED IN THE $K_{\scriptscriptstyle D}$ TESTS

Please provide a discussion of the selection of groundwater sources used in the K_d tests. The August 3, 1995 report used a blend of groundwater samples from three wells near the LARW cell while the January 25, 1996 report used groundwater from only one well near the 11(c)(2) cell. Explain the rationale for the different groundwater sample selections for the two sets of tests. Also, explain or justify why the groundwater samples are representative or appropriate for the site-specific K_d measurements. Details in this justification should include, but are not limited to, the groundwater pH, Eh, and major ion content.

The groundwater used during the testing reported in August 1992 was taken from wells on three sides of the LARW. These three wells are located on three sides of the LARW and are established monitoring wells for that facility. In that light, it is difficult to envision how they could be more representative of groundwater conditions associated with the LARW. Table 1 below reports the average major ion composition of the groundwater at GW-20, GW-22 and GW-64 (the wells sampled for the K_d program reported in the August 1995 Bingham Environmental report) as well as the analysis of the stock groundwater used for the K_d study (analyzed by Barringer Labs). By inspection, it can be seen that there is not a wide range of compositions to be considered. GW-25 was sampled for the K_d work performed and reported in the January 1996 Bingham Environmental report and is also shown in Table 1 and can be seen to be consistent with all other samples. GW-25 was sampled for the K_d work performed and reported in the January 1996 Bingham Environmental report and is also shown in Table 1 and can be seen to be slightly more saline than the other groundwater used. As sorption is commonly observed to diminish with increasing salinity, the K_d values obtained using water from GW-25 would tend to produce slightly lower values and, with respect to modeling transport, err on the side of environmental protection.

Table 1. Average groundwater composition associated with the LARW.

CONSTITUENT*	GW-20	GW-22	GW-64	GW-25	TEST (8/95)
Calcium	412.5	412.5	280	528	400
Magnesium	685	625	385	878	605
Potassium	527.5	497.5	385	585	503
Sodium	15250	14750	11250	16500	15400
Alkalinity	207.5	322.5	220	197	243
Chloride	23500	22750	18000	24000	23300
Sulfate	4175	2400	1700	5075	2550
TDS	46250	44000	33250	50250	41500
pН	7.6	7.6	7.6	7.5	7.52
Redox(Eh)	NA	NA	NA	NA	160

^{*} all concentrations as mg/L except pH, su, and redox, mV; NA- not available

Eh measurements have been shown repeatedly to be unreliable using a platinum electrode in solutions that have do not have active redox couples at meaningful concentrations (e.g. Fe^{2+}/Fe^{3+} at concentrations on the order of 10^{-4} mol/L, 5-6 ppm). The groundwater at the Envirocare site does not appear to have suitable characteristics to make Eh measurements meaningful. Further, past field measurements of Eh have been very erratic and tend to support the hypothesis that the solutions are not well poised (the electrochemical equivalent of the acid-base concept buffered). Thus, Eh is not suitable for assessing the representativeness of solutions used for testing. Measurements of dissolved oxygen (DO) would probably more suitable for assessing the oxidation potential of solutions, but were not performed as part of the K_d study and have not previously been measured in the field. Do measurements are currently being collected at the site.

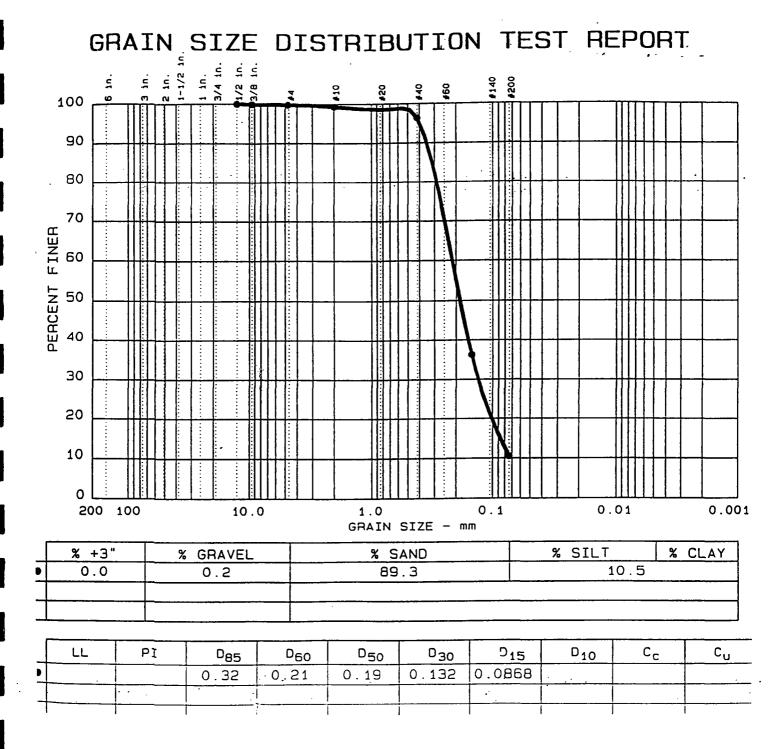
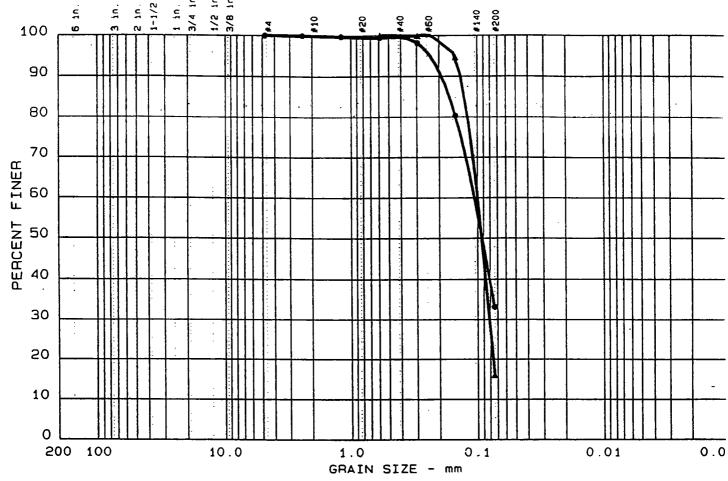


Figure 2. Grainsize distribution of test sands.

GRAIN SIZE DISTRIBUTION TEST REPORT

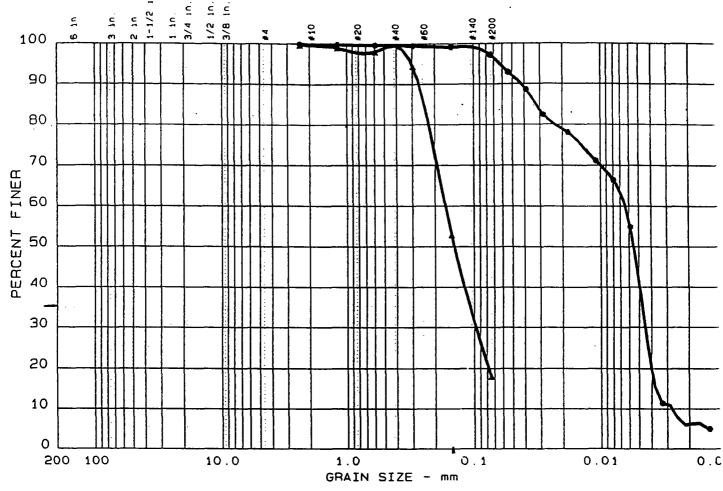


	Test	%+75 mm	% GRAVEL	% SAND	% SILT	% CLAY
•	5	0.0	0.0	66.7	33.3	
A	6	0.0	0.0	83.8	16.2	

	LL "	. PI	D _{85.}	D ₆₀	D ₅₀	D ₃₀	D ₁₅ .	D ₁₀	. C _c	Cu
•	N/A	N/A	0.17	0.11	0.09					•
A	N/A	N/A	0.13	0.10	0.09	0.081				

Figure 3. Grainsize of Unit 3 sand, well 19A.

GRAIN SIZE DISTRIBUTION TEST REPORT



	Test	%+75 mm	% GRAVEL	% SAND	% SILT	% CLAY
•	12	0.0	0.0	2.8	56.2	41.0
lacksquare	9	0.0	0.0	82.0	18.0)

	LL.	PI	D ₈₅	D ₆₀	D ₅₀	D ₃₀	. D ₁₅	D ₁₀	Сс	Cu
•	44	18			0.01	0.004	0.0037	0.0027	1.16	2.3
	N/A	N/A	0.24	0.17	0.14	0.097				

Figure 4. Grain size distribution of Unit 3 sand.

3. SOIL-TO-WATER RATIO USED IN THE K_D TESTS

The soil-to-water ratio used in the K_d tests was approximately 1:4 instead of the 1:1 ratio expected under field conditions. Apparently, the 1:4 ratio was used because it is part of the ASTM test procedure. Please discuss the effects on the test results of using a 1:4 ratio versus using a 1:1 ratio.

Unless active precipitation of a solute occurs, the measurement of K_d is not effected by the water rock ratio, although the % reduction in solute concentration may appear greater. The value of K_d is the ratio of the concentration of an analyte on a solid phase compared to the coexisting concentration in the solution contacting the solid.

$$K_d = C_{solid} / C_{solution}$$

where C solid has units of mg/kg and C_{solution} mg/L. If higher proportions of solid are used relative to the same amount of test solution, more mass of a solute may be taken up, but as that mass is divided by the mass of solid onto which the sorption occurs, the concentration term would be expected to remain fixed relative to a coexisting solution concentration.

If a chemical reaction occurs between the test solution and solid that causes precipitation reaction, from either a change in pH or redox potential, the extent of this effect is lessened with a higher water rock ratio and produces a more reliable measurement of K_d . Thus, using a soil-to-water ratio of 1:4 would be more appropriate than lower ratios.

4. INFORMATION ON THE $K_{\scriptscriptstyle D}$ TEST CONDITIONS

Please provide information on the K_d test conditions and sample handling procedures to help demonstrate whether the test conditions mimic field conditions. Discuss the use of preservatives (if any) in groundwater samples, and the laboratory test conditions (.e.g., temperature, Eh and pH contact solutions, major chemical constituents of the contact solutions, an chemical forms of radionuclides in contact solutions).

Redox and pH measurements of contact solutions were taken prior to contact and following equilibration. In general, pH redox conditions increased slightly during testing. No preservatives were added to the sampled groundwater, as is evident by the neutral pH maintained, the same condition as observed in the field setting. The results are reported in Table 2

Table 2. Redox pH conditions for K_d tests.

Nuclide	PH _{INIT}	EHINIT	PH_{FINAL}	EH _{FINAL}
C ¹⁴	7.52	160	7.73	216
I^{129}	"	66	7.82	205
Np ²³⁷	"	66	7.81	206
Tc ⁹⁹	"	66	7.83	218
U-nat	"	66	7.61	204

The changes reported in Table 2 are not great and initial and final conditions are considered to be consistent. As the introduction of the standards used in the testing will themselves be expected to exert some influence on the solution, it would be difficult to constrain the experiment any better.

The extent to which groundwater samples collected for laboratory study may have increased their DO concentration to perturb the measurement of K_d is limited. Specifically, water saturated with oxygen at atmospheric pressures will have on the order of 8-9 ppm DO. The concentration will not be higher than this unless the solution is vigorously agitated, but even then will settle at 8-9 ppm on standing. Solutes tested for adsorption which have only one expected oxidation state (C^{14} , I^{129} and T^{29}) will be unaffected by the presence of oxygen in test solutions. Others that have more then one potential oxidation state (U-nat and Np^{237}) will be unaffected if the form used during testing was the highest oxidation state. In the K_d study, U-nat was in the +6 oxidation state, which is expected since this is the soluble form of the element and, thus, uranium would not be affected by the introduction of oxygen to test solutions. Np^{237} used in the K_d determinations was in the +4 oxidation state, while +5 is possible. The lower oxidation state is soluble only at low pH (≤ 2). Thus, for the assumed condition of depleted dissolved oxygen and neutral pH in the groundwater associated with the site (low Eh reducing conditions), the +4 oxidation state of Np^{237} would be anticipated, but owing to its relatively insoluble nature, would not persist in the solution phase and would be precipitated.

5. LINEAR RELATIONSHIP BETWEEN SOIL AND WATER CONCENTRATIONS

The assumed linear relationship between soil and water concentrations has generally not been demonstrated by the K_d tests. Please provide additional discussion and test data to help support the linear hypothesis. Also, justify that the radionuclide concentrations used in the contact solutions bound the range of expected leachate concentrations in the disposal cell.

A linear relationship exists between soil and water concentrations for some elements and not for others. Np²³⁷ (with the deletion of one point), C¹⁴, Tc⁹⁹ are well described by a linear relationship (see Figure 5-7). Uranium appears as well to be fit by a linear relationship (Figure 8), although this is a little misleading due to only having data for one concentration. I¹²⁹ is less well fit by a linear relationship

(Figure 9). Any deviations for any nonlinearity (e.g. I¹²⁹) are not satisfactorily fit by other adsorption isotherms such as the Langmuir or Freundlich and, thus, while having associated uncertainty, a linear relationship appears suitable for the present data.

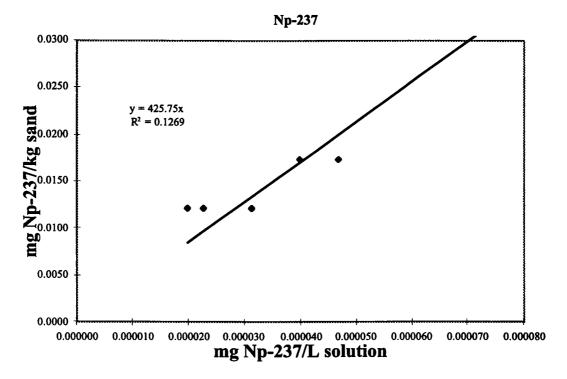


Figure 5. Np²³⁷ isotherm.

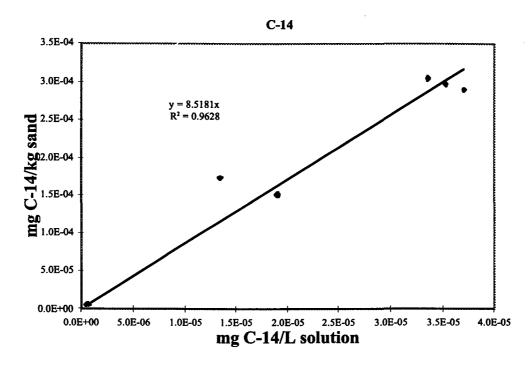


Figure 6. C¹⁴ isotherm.

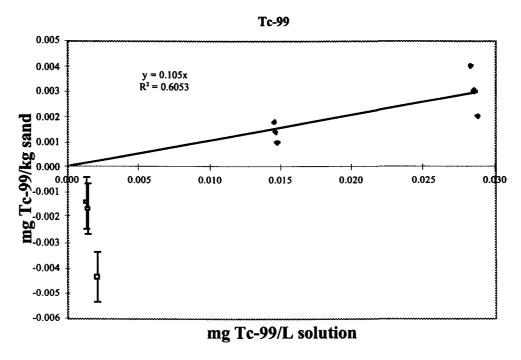


Figure 7. Tc^{99} isotherm.

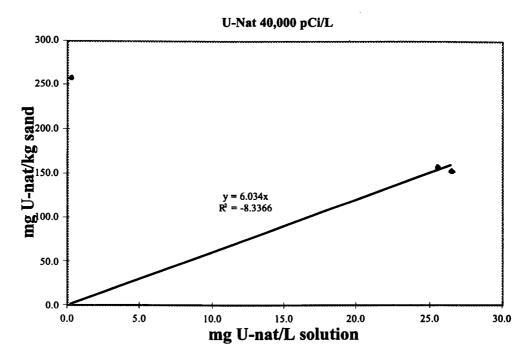


Figure 8. U-nat isotherm.

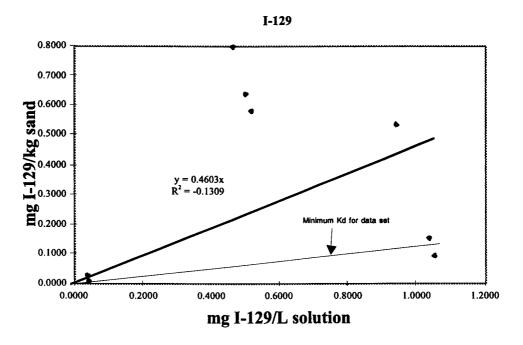


Figure 9. I¹²⁹ isotherm.

Note that this response to UDEQ interrogatories has reanalyzed the experimental data in the proper fashion by determining the K_d for each element by regressing the concentration on the solid against the concentration in the solution. Only Tc^{99} has data points which plot as negative values on the solid concentration axis. By definition, these points are erroneous as Tc is a synthetic element and is not present in the solids associated with the site. The error associated with the analysis of each of these points is shown in the figure and demonstrate that not even the analytical error is sufficient to correct these data points. There is probably simply an analytical error associated with the low concentrations of these points. When valid Tc^{99} data are regressed to determine the slope (the actual definition of K_d) the value obtained is 0.11 (to two significant figures), higher than previously calculated using an averaging technique for K_d 's calculate for individual points, which erroneously included the low concentration data. Table 3 reports the originally calculated K_d 's for all nuclides and recalculated values using linear regression .

Table 3. Current and recalculated K_d values.

RADIONUCLIDE	CURRENT VALUE	PROPOSED CORRECTED VALUE
C ¹⁴	9.0	8.52
I ¹²⁹	0.7	0.46
Np ²³⁷	400	425
Tc ⁹⁹	0.07	0.11
U-nat	6.0	6.0

The corrected values reported above represent the slope of the graphs of concentration in the solid versus concentration in the solution. All elements demonstrated linearity and thus the simple least squares fit produces an appropriate K_d from the slope. I^{129} , which has the lowest degree of linearity associated with the regression, yields a K_d of 0.46. However, this value could be as low as that associated with shallowest slope passing through the scatter of data, a K_d of 0.12.

Although the K_d value recalculated for Np^{237} is derived from a linear relationship, the large error associated with analysis of test solutions cast uncertainty on the K_d determination. However, the values obtained are consistent with sorption behavior observed for neptunium for a variety of materials over a range of pH and salinity solution conditions. The figure below graphs available data for neptunium sorption from the literature. It is clear that there exists a general correlation between K_d and the pH of solution for all materials and that the minimum literature K_d , currently proposed for modeling, is at the extreme low end of the pH range. Specifically, the literature low K_d was measured at a pH of 2.7, inconsistent with the site groundwater pH of about 7.5 and is thus not suitable. At a pH of 7.5, the general trend of the data suggest a minimum K_d value of about 8. It is significant to note that the general trend represented in Figure 10 established by a range of solid materials and also includes measurements in

brines. Thus, while the site specific measurement of $K_d = 400$ may be suspect in terms of analytical concerns, it is remarkably consistent with literature data and would appear to be more representative of site conditions than the low literature value which was measured under very acidic conditions.

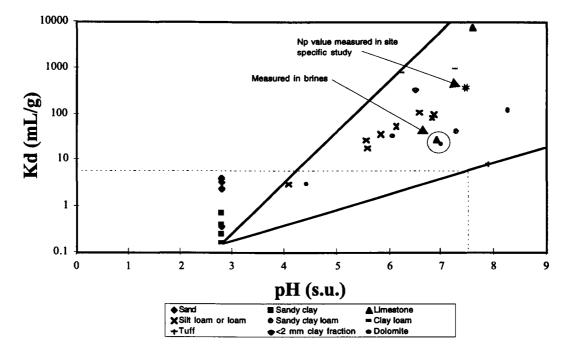


Figure 10. Literature K_d values for neptunium.

The extent to which concentrations used to make laboratory determinations are consistent to future field conditions is not readily documented. It is not possible to predict a priori the concentrations from a waste stream whose specific composition is unknown. However, concentrations used in the experimental program were derived using a "guessed" K_d and possible source concentration to estimate potential concentrations in leachates. However, with the reasonably demonstrated linearity of the isotherms, it is not a critical concern that exact concentrations, or even bounding concentrations, for solutes be determined. That is the strength of using a linear isotherm.

6. FORMATION OF A PRECIPITATE IN THE CONTACT SOLUTIONS

The K_d tests for uranium, carbon, and technetium noted the formation of a precipitate in the contact solutions. Provide information on the possible chemical composition of the precipitates and their effects on the test results. The information on the chemical composition of the contact solutions, requested in item 4 above, will help support the conclusions on precipitate composition. Please indicate if the precipitation reaction is possible or likely to occur under field conditions.

Precipitates formed during the preparation of contact solutions due to either salinity or pH changes associated with introduction of the purchased radionuclide standard into site groundwater. Potential identities for uranium precipitates includes autinite, a common, yellow colored uranium phosphate (Ca(UO₂)₂(PO₄)₂ 10-12 H₂O). C¹⁴ (as benzoic acid, C₆H₅COOH) may have precipitated as a salt of benzoate, C₆H₅COOX, where x may be Ca, Mg, etc. A detailed literature review of the solubility of benzoate salts as a function of salinity has not been performed. Little thermodynamic data are readily available for technicium to determine what solid phases may have precipitated.

To the extent that leaching reactions in placed waste can produce solutions like the chemical standards used to prepare experimental solutions during K_d testing, the precipitation reactions could and would occur in the field setting when those leachates mixed with groundwater. It seems unlikely that the commonly low pH conditions of chemical standards (U-nat) would be reproduced in placed waste, nor would it be expected that all C^{14} would be released from placed waste as benzoic acid. Hence the observations of precipitation made during lab testing would not be expected in the field.

7. NEPTUNIUM STANDARD USED IN THE TEST

The August 3, 1995 report, Appendix D, notes that the test results for neptunium are invalid because of a problem with the neptunium standard used in the test. Please provide additional testing for neptunium or adopt the previous neptunium K_d (minimum from the literature).

The neptunium standard used during K_d testing was not flawed, it was simply a lower concentration than the lab personnel expected. Hence, for the tests reported, the lab experimental program used concentrations lower than thought likely when the program was established. However, to the degree that a linear relationship exists, using a lower initial concentration leads to an identical determination of K_d . Thus, there is no need, on the basis of a "flawed" standard to repeat determinations of Np^{237} K_d .

The lower concentrations produced for testing, because of the low concentration standard can lead to increased analytical errors (due to proximity of the detection limit for the method), but in no way would be expected to make the concept of measuring K_d invalid.

8. POSSIBLE EFFECTS OF COMPETING IONS IN THE $K_{\scriptscriptstyle D}$ TEST

Provide information on the possible effects of competing ions in the K_d test contact solutions, including the effects of ions in the saline groundwater. This information must include an evaluation of the competing ion effect on contaminants previously considered to be non-mobile (e.g., radium).

The presence of dissolved solids can lead to K_d values that are less than those determined for relatively more dilute solutions. With respect to the elements tested, the K_d values determined would be consistent with conditions expected to persists in the groundwater environment. These K_d values would produce lower concentrations of chemicals leached from placed waste during PATHRAE modeling, as the salinity of fluids percolating through this waste would not exceed, and probably be lower, than the salinity of local groundwater.

9. SPECIATION OF RADIOACTIVE CHEMICALS

Please provide information on the speciation of radioactive chemicals in the contact solutions. As the radionuclides absorb onto the solids, their chemical forms may change from the initial compounds in the contact solutions. Precipitates may also form and remove radionuclides from solution, leading to higher effective K_d values. The speciation information should include consideration of the precipitates that were formed in the uranium, carbon, and technetium tests.

The radioactive compounds used during attenuation testing are reported in Table 4.

Table 4. Radionuclide speciation used in K_d testing.

RADIONUCLIDE	Compound	OXIDATION STATE
C ¹⁴	benzoic acid, C ₇ H ₆ O ₂	+4
I ¹²⁹	sodium iodide, NaI	-1
Np ²³⁷	neptunium nitrate, Np(NO ₃) ₄	+4
Tc ⁹⁹	ammonium technetate, (NH ₄)TcO ₄	+7
U-nat	U in water	+6

Precipitates were formed during attenuation testing, but do not impact the distribution ratios determined because these precipitates were filtered from the solutions prior to soil contact. The formation of precipitates during soil contact does not appear to have occurred, as the analyst's observations, included in the Bingham Environmental report, indicate that all solutions were clear.